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Regional versus local drivers of water quality in the Windermere catchment, Lake District, United Kingdom: The dominant influence of wastewater pollution over the past 200 years

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Abstract

Freshwater ecosystems are threatened by multiple anthropogenic stressors acting over different spatial and temporal scales, resulting in toxic algal blooms, reduced water quality and hypoxia. However, while catchment characteristics act as a 'filter' modifying lake response to disturbance, little is known of the relative importance of different drivers and possible differentiation in the response of upland remote lakes in comparison to lowland, impacted lakes. Moreover, many studies have focussed on single lakes rather than looking at responses across a set of individual, yet connected lake basins. Here we used sedimentary algal pigments as an index of changes in primary producer assemblages over the last ~200 years in a northern temperate watershed consisting of 11 upland and lowland lakes within the Lake District, United Kingdom, to test our hypotheses about landscape drivers. Specifically, we expected that the magnitude of change in phototrophic assemblages would be greatest in lowland rather than upland lakes due to more intensive human activities in the watersheds of the former (agriculture, urbanization). Regional parameters, such as climate dynamics, would be the predominant factors regulating lake primary producers in remote upland lakes and thus, synchronize the dynamic of primary producer assemblages in these basins. We found broad support for the hypotheses pertaining to lowland sites as wastewater treatment was the main predictor of changes to primary producer assemblages in lowland lakes. In contrast, upland headwaters responded weakly to variation in atmospheric temperature, and dynamics in primary producers across upland lakes were asynchronous. Collectively, these findings show that nutrient inputs from point sources overwhelm climatic controls of algae and nuisance cyanobacteria, but highlights that large-scale stressors do not always initiate coherent regional lake response. Furthermore, a lake's position in its landscape, its connectivity and proximity to point nutrients are important determinants of changes in production and composition of phototrophic assemblages.

KEYWORDS

algal assemblages, climate change, eutrophication, landscape, multiple stressors, synchrony, wastewater

1 | INTRODUCTION

Globally, inland waters have been subject to diverse biogeochemical alterations and biodiversity declines since the Industrial Revolution due to greenhouse gas emissions, pollution and food-web modification (Wolfe et al., 2013). Historical changes were particularly notable after World War II (ca. 1945), in the period known as the “Great Acceleration” (Steffen, Crutzen, & McNeill, 2007). Consequently, there have been considerable declines in the water quality and biological integrity of surface waters in both populated and remote regions (Vörösmarty, McIntyre, & Gessner, 2010). Furthermore, with the fourfold increase in global population, doubling of agricultural activity, and ninefold greater water use of the 20th century expected to increase further in the next 30 years (McNeill, 2000), there is a profound need to better understand the hierarchical relationships among human causes of ecosystem disturbance and how this is modified by lake and catchment characteristics. This novel study compares long-term change in assemblages of primary producers in diverse lakes (headwaters to terminal basins) within a heterogeneous landscape to assess the hierarchy of relationships among drivers of ecological modifications over the last ~200 years.

In principle, environmental parameters and anthropogenic stressors affect surface waters on diverse spatial scales (Soranno, Webster, Cheruvilil, & Bremigan, 2009). As described in the *Em-flux* framework of Leavitt et al. (2009), environmental variation is transmitted to lakes by inputs of energy (*E*) (irradiance, heat, kinetic *E* of wind) or mass (*m*) (water, nutrients, suspended materials). For example, regional variation in solar insolation arising from changes in climate systems (e.g. El Niño-Southern Oscillation; Pacific Decadal Oscillation) result in temporally coherent changes in lake parameters such as dates of ice melt, thermal stratification and annual plankton development (Dröscher, Patoine, Finlay, & Leavitt, 2009). Similarly, broad-scale shifts in land use practices across catchments of similar geomorphology and topography may also initiate coherent changes in lakes with common histories of agricultural development (Anderson, Bennion, & Lotter, 2014; Keatley, Bennett, Macdonald, Taranu, & Gregory-Eaves, 2011). However, the magnitude and synchrony of lake responses to regional environmental forcings, such as climate, will also depend on basin and catchment characteristics, such as elevation, size, depth and geology (Leavitt et al., 2009). Acknowledging the importance of catchment characteristics as “filters” of multiple stressors (*sensu* Blenckner, 2005) can help detect sites more vulnerable to stressor effects than others (Leavitt et al., 2009; Mills et al., 2017).

In contrast, variation in mass (water, nutrients, suspended matter) influx to lakes arises from urban pollutants such as Wastewater Treatment Works (WwTWs) (Jenny, Normandeau, & Francus, 2016),

short-term or site-specific variation in precipitation and inflow regimes (Chen, McGowan, Xu, Zeng, & Yang, 2016), or basin-specific changes in agricultural practice (Maheux, Leavitt, & Jackson, 2016), all of which influence delivery of particulate material (Dearing & Jones, 2003). Such local disruptions often reduces synchrony of limnological variables among basins (Leavitt et al., 2009; Maheux et al., 2016) and impede the effectiveness of generalized mitigation strategies. The development of effective management of aquatic resources would benefit from a hierarchical understanding of the relative importance of regional versus local stressors that threaten freshwaters (Jackson, Woodford, & Weyl, 2016; Vörösmarty et al., 2010).

Landscape-scale palaeolimnology is a promising approach to study the unique and interactive effects of multiple stressors on lakes over relevant timescales (Anderson, 2014; McGowan & Leavitt, 2009; Taranu et al., 2015). In large flat watersheds, this approach shows that short-term variation in nutrient influxes to lakes associated with agricultural development can override the effects of long-term variation in climate (Leavitt et al., 2009; McGowan & Leavitt, 2009; Patoine & Leavitt, 2006). In contrast, retrospective studies of remote mountain catchments suggest that upland basins are more sensitive than lowland lakes to regional, longer term, climate variability (i.e. temperature) due to greater temperature fluctuations with altitude and distance from localized anthropogenic disturbance (Lami et al., 2010). Despite these observations, few studies to date have explicitly quantified the relative importance of climate and anthropogenic mechanisms across broad temporal scales and diverse lake characteristics (Soranno et al., 1999).

Here, we compare changes in abundance and composition of phototrophic assemblages in 11 lakes within a single watershed to determine the influence of regional and local stressors over the last ~200 years. Basins range in elevation from 39 m.a.s.l. (Windermere terminal basins) to 469 m.a.s.l. (Stickle Tarn), which allows comparisons between upland lakes (>100 m.a.s.l.) remote from human disturbance, and lowland (<100 m.a.s.l.) lakes that are more directly affected by anthropogenic processes. Our study region, the Windermere catchment of northwestern England (Figure 1), is typical of many inhabited temperate Northern Hemisphere watersheds. This network of glacially derived lakes has been subject to substantial anthropogenic perturbation (McGowan et al., 2012), and marked climate change in recent centuries (Dong et al., 2012), resulting in substantial water quality issues (Pickering, 2001) including nuisance cyanobacterial blooms (Elliott, 2012). In this study, we tested three hypotheses relevant to historical ecological change during the past 200 years: (a) The magnitude of phototrophic assemblage modification is greater in lowland than upland lakes due to more intensive human activities (agriculture, urbanization). (b) Regional parameters,

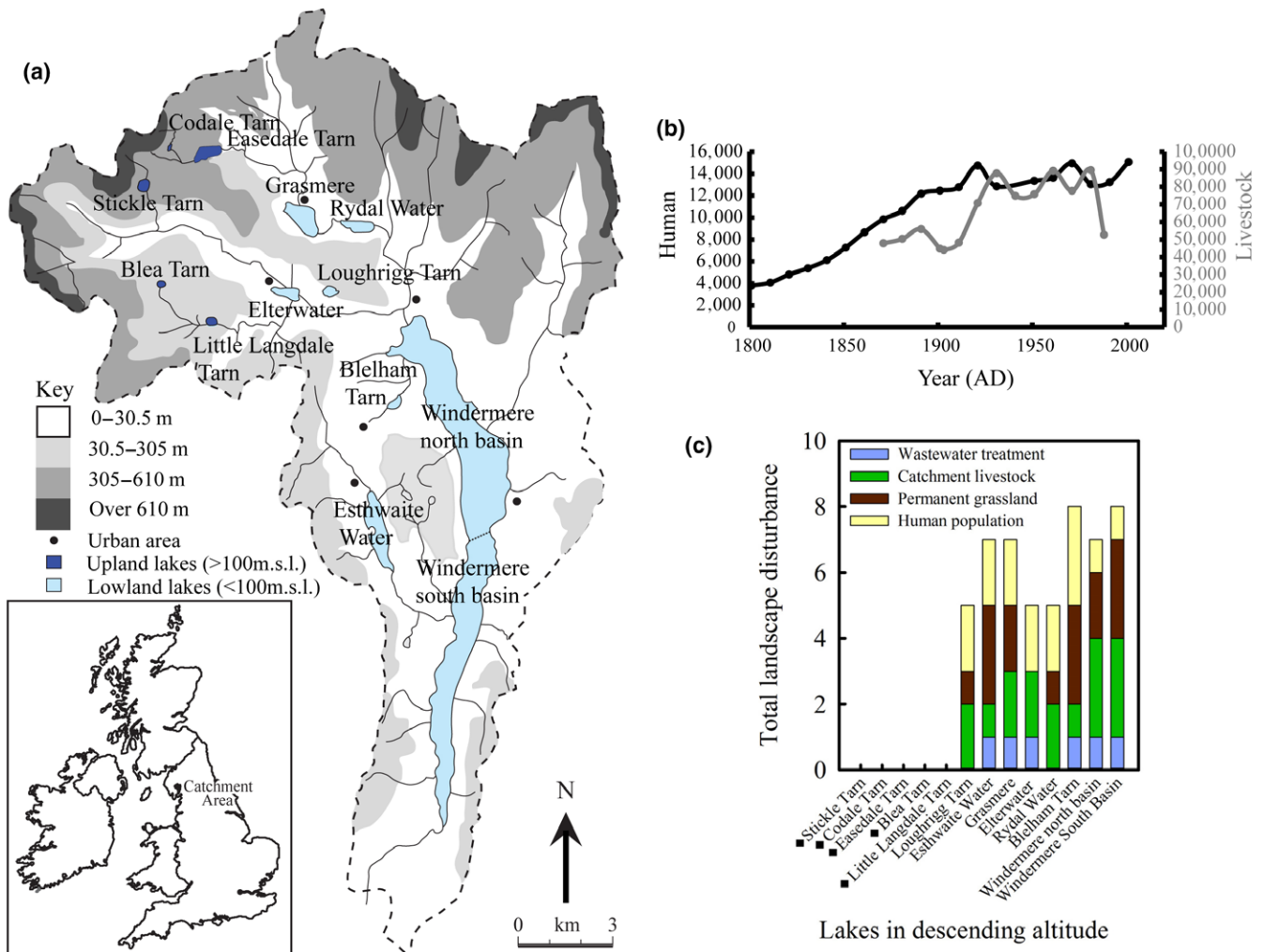


FIGURE 1 (a) Location of study lakes in the Windermere catchment (modified from McGowan et al. (2012) and Pickering (2001)). (b) Temporal changes in human (number of persons; black) and livestock (total number of cattle, sheep, pigs, horses and poultry; grey) numbers in the catchment indicate intensification of human activities from 1800 onwards. (c) Spatial patterns of human disturbance indicators arranged in order of declining altitude (upland lakes >100 m.a.s.l. indicated by squares). Total landscape disturbance = Wastewater Treatment Work in lake catchment (presence = 1) + historical catchment livestock densities (mean individuals per hectare from 1866 to 1988 (<1 ind/ha = 0, >1 ind/ha = 1, >3 no/ha = 2)) + % permanent grassland (mean from 1866 to 1988 (<30% = 0, >30% = 1, >40% = 2, >50% = 3)) + catchment human population human densities (mean individuals per hectare from 1801 to 2001 (<0.01 ind/ha = 0, >0.01 ind/ha = 1, >0.1 ind/ha = 2, >0.3 no/ha = 2))

such as climate, are the predominant factors regulating lake primary producer communities in upland headwater lakes, with local human activities explaining modifications in the lowlands (e.g. Wastewater Treatment Works; WwTWs). (c) Change in primary producer communities is more spatially coherent in upland than lowland basins, reflecting asynchronous basin-specific effects of human activities in the lowlands.

2 | MATERIALS AND METHODS

2.1 | Study area

The Windermere catchment (54°18' N; 2°54' E) is situated in the Lake District National Park in northwest England, United Kingdom, and comprises 11 lowland and upland lakes which feed into Windermere,

England's largest twin-basin lake (Figure 1; Table 1). Receding glaciers formed lakes in the catchment at the end of the last glacial period, resulting in variable morphology of individual subcatchments. Regional climate is classified as temperate oceanic (Cfb in Köppen system) (Peel, Finlayson, & McMahon, 2007), with generally cool temperatures (mean high 13°C, mean low 5.8°C) and high average annual precipitation (>1,500 mm/year), but wide spatial and temporal variation (up to 5,000 mm/year). The shading effect of the Lake District mountains reduces temperatures and sunlight in the uplands, and rainfall is predominantly orographic, with higher mean annual precipitation in upland areas compared to lowland areas (Chiverrell, 2006; George, Hurley, & Hewitt, 2007; Kenworthy, 2014; Met Office, 1971).

The Windermere catchment is composed of two distinctive geological areas with hard, slow-weathering extrusive lavas of the Borrowdale Volcanic Group (BVG) in the north separated from Silurian

TABLE 1 Physical and limnological characteristics of lakes in the Windermere catchment in order of descending altitude

Site	Geology	Mean depth (m)	Max depth (m)	Length (km)	Area (km ²)	Catchment area (km ²)	Catchment to lake area ratio	Altitude (m.a.s.l.)	Mean catchment altitude (m.a.s.l.)	Mean Retention time (days)	TP (mg/L) or trophic status	Primary data source for physical and limnological variables
Stickle Tarn	BVG	8	14	0.37	0.083	1.8	21.7	469	610	39	11.7 ^a	8, 9 & 11
Codale Tarn	BVG	3.3	7	0.2	0.013	0.40	30.8	467	610	12	10.5 ^a	8, 9 & 11
Easedale Tarn	BVG	10.5	22.5	0.48	0.106	2.7	25.5	279	510	55	11.3 ^a	8, 9 & 11
Blea Tarn	BVG	7	8	0.26	0.038	1.16	30.5	192	520	-	13.3 ^a	1 & 11
Little Langdale Tarn	BVG	2.7	9.5	0.375	0.065	12	184.6	102	520	3.3	14.6 ^a	8 & 11
Loughrigg Tarn	BVG	6.9	10.3	0.4	0.07	0.95	13.6	94	175	117	23.5 ^a	11 & 14
Esthwaite Water	SIL	6.4	15.5	2.5	1	17.0	17	65	148	100	28 (mean for 2008)	13, 14 & 15
Grasmere	BVG	7.74	21.5	1.6	0.644	30.2	46.9	62	328	25	Eutrophic	2, 3, 12 & 13
Elterwater inner basin	BVG	2.3	7.4	1	0.036	1.0	27.9	55	108	106	Eutrophic	4, 6, 10, 13 & 14
Elterwater middle basin	BVG	2.3	6	1	0.074	1.0	13.5	55	108	26	Mesotrophic	6, 5, 13 & 14
Elterwater outer basin	BVG	2.5	7.5	1	0.083	1.0	12	55	108	20	Oligotrophic	6, 13 & 14
Rydal Water	BVG	4.4	19	1.2	1.5	33	22	53	312	9		13 & 14
Blelham Tarn	SIL	6.8	14.5	0.67	0.1	4.0	40	42	105	50	Eutrophic	13, 14 & 16
Windermere north basin	SIL	25.1	64	7	8.1	175	21.6	39	231	180	Mesotrophic	7 & 17
Windermere south basin	SIL	16.8	42	9.8	6.7	200	37.3	39	231	100	Eutrophic	7 & 17

BVG, Borrowdale Volcanic Group; SIL, Silurian flags and slates.

^adenotes surface water total phosphorus (TP) samples collected and measured in July 2013. Grey shading denotes upland lakes >100 m.a.s.l.Primary data references: ¹Haworth (1969), ²Hall, Collins, Jones, and Horsley (1978), ³Reynolds and Lund (1988), ⁴Carvalho and Moss (1995), ⁵Beattie et al. (1996), ⁶Zinger-Gize, Hartland, Saxby-Rouen, and Beattie (1999), ⁷Reynolds and Irish (2000), ⁸Tipping et al. (2000), ⁹Tipping et al. (2002), ¹⁰Goldsmith et al. (2003), ¹¹Haworth et al. (2003), ¹²Barker et al. (2005), ¹³George et al. (2007), ¹⁴Maberly et al. (2011), ¹⁵Dong et al. (2012), ¹⁶Foley, Jones, Maberly, and Rippey (2012), ¹⁷McGowan et al. (2012).

Coniston Flags and Slates (SIL) to the south by a thin band of Coniston Limestone. SIL rocks are easily weathered resulting in surface waters with higher ionic concentrations and buffering capacity than the BVG group, where overlying waters have a similar ionic composition to precipitation (Sutcliffe et al., 1982; Thornton & Dise, 1998). These differences in geology influence soil cultivability and have been described as the 'Pearsall sequence' of lake evolution with 'primitive' lakes overlying BVG geology and 'evolved' SIL lakes having distinct differences in lacustrine communities (Pearsall, 1921). Most of the catchment lakes are predominantly phosphorus (P) limited, although production in some of the oligotrophic upland lakes is colimited with nitrogen (Maberly, King, Dent, Jones, & Gibson, 2002).

Distinct differences in land use and lake trophic status exist between cultivated and uncultivated subcatchments in the Windermere region (George, Talling, & Rigg, 2000). Upland cover is largely grassland for rough grazing of sheep and limited cattle farming, although some native woodland exists to the west of Lake Windermere (Maberly et al., 2003; Pickering, 2001). In lowland areas, agricultural intensification in the latter half of the 20th century improved pasture conditions to facilitate increased livestock densities in the fertile lowland subcatchments of Esthwaite Water and Blelham Tarn (Heathwaite, Johnes, & Peters, 1996). Furthermore, tourism is an important industry (15.8 million visits per year), particularly in the larger urban settlements of Ambleside and Bowness-on-Windermere on the shores of Lake Windermere (Harvey, Thompson, Scott, & Hubbard, 2013), and has resulted in wastewater discharge into, and eutrophication of Windermere, Grasmere, Elterwater and Esthwaite Water (McGowan et al., 2012; Talling, 1999). This spatial variability in land use and geology leads to pronounced differences in modern lake chemistry and ecology, despite their close proximity (Talling, 1999).

2.2 | Sediment coring

Lake sediment cores were retrieved from the deepest point of 11 lakes in the Windermere catchment from November 2011 to July 2013 using a 50-cm-long Glew gravity corer (Glew, 1988) for the unproductive upland tarns (Stickle Tarn, Codale Tarn and Easedale Tarn), a 1-m mini-Mackereth corer (Mackereth, 1969) for the more productive lowland sites (Blelham Tarn, Esthwaite Water, Rydal Water) and a 1-m HON-Kayak gravity corer (Renberg, 1991) for the remaining basins (Blea Tarn, Little Langdale Tarn, Loughrigg Tarn, Grasmere, Elterwater inner, middle and outer basins). Cores from the north and south Windermere basins were collected in 2009 (see McGowan et al., 2012). All cores were sectioned into 0.5-cm intervals within 48 hr of collection, sealed in airtight plastic bags, and samples frozen (-20°C) for pigment analysis or refrigerated for other analyses.

2.3 | Sediment chronology

Freeze-dried sediments (48 hr, 0.1 Pa) from Blelham Tarn, Esthwaite Water, Rydal Water, Easedale Tarn and Stickle Tarn were dated using ^{210}Pb , ^{226}Ra via its daughter isotope ^{214}Pb , ^{137}Cs and ^{241}Am

by direct gamma assay. Analyses were conducted using an ORTEC HPGe GWL series, well-type coaxial, low background, intrinsic germanium detector at the Environmental Radiometric Facility at University College London, United Kingdom. Freeze-dried sediments from Elterwater inner basin, Grasmere, Loughrigg Tarn and Codale Tarn were dated using ^{210}Pb , ^{137}Cs and ^{214}Pb activities using a well-type, ultralow background HPGe gamma ray spectrometer at the Aquatic Research Centre, University of Brighton. Longer count times were used for these four cores due to the high water content and flocculent nature of the sediment (e.g. $>400,000$ s for Elterwater inner basin).

The constant rate of supply model (CRS) was used to estimate sediment ages for Blelham Tarn, Codale Tarn, Easedale Tarn, Esthwaite Water, Loughrigg Tarn, Rydal Water and Stickle Tarn, reflecting the nonmonotonic features of the ^{210}Pb profiles (Appleby, 2001). All chronologies were validated based on ^{137}Cs and ^{241}Am profiles. However, further validation was required for Elterwater inner basin and Grasmere due to the very low activity of ^{210}Pb in the sediments, which prevented construction of a meaningful age-depth model. At these two sites ^{137}Cs profiles with distinct ^{137}Cs maxima from the 1986 Chernobyl accident and 1963 atmospheric atomic weapons testing maxima were used to estimate ages. The depth where ^{137}Cs activity reached zero was assigned as 1940 prior to global nuclear weapons testing (Ritchie & McHenry, 1990). The chronology was interpolated using these dates and that of core collection. These chronologies were then further validated by comparing proxies to previous studies at Grasmere (Barker, Pates, Payne, & Healey, 2005) and Elterwater inner basin (Lund, 1981), including loss-on-ignition at 550°C (%LOI₅₅₀), diatom (diatoxanthin) and green algal (lutein) pigment concentrations, and morphological fossils from diatoms/chlorophytes. Linear interpolation using the last two deepest age-depths to extend back to 1800 was undertaken for cores whose deepest age-depth was before this date (Grasmere, Elterwater inner basin and Windermere's north basin). Cores from Blea and Little Langdale Tarns, Elterwater middle and outer basins were not dated due to lack of funding and were excluded from analyses where absolute dates were required. These latter sites were included in the Mann-Kendall trend analyses (described below) to help interpret catchment-wide concentration trends, but their undetermined time series must be acknowledged.

2.4 | Sedimentary pigment analysis

Sedimentary carotenoids were used to reconstruct changes in abundance and composition of phototrophic assemblages of algae and cyanobacteria (Leavitt & Hodgson, 2001). Pigments from aliquots of freeze-dried samples were extracted, solutions filtered, and individual compounds then separated and quantified using an Agilent 1,200 series quaternary pump, autosampler, ODS Hypersil column (250×4.6 mm; $5 \mu\text{m}$ particle size) and photo-diode array (PDA) detector as previously described (Chen, Bianchi, McKee, & Bland, 2001). Pigments were expressed as nmole pigment/g organic matter (OM) using %LOI₅₅₀ as a determinant of the OM fraction (Heiri, Lotter, & Lemcke, 2001). The HPLC was calibrated using commercial

pigment standards from DHI (Denmark). Analysis included chemically stable, taxonomically diagnostic carotenoids representing total algal production (β -carotene), cryptophytes (alloxanthin), potentially N_2 -fixing cyanobacteria (aphanizophyll), canthaxanthin (colonial cyanobacteria), zeaxanthin (all cyanobacteria), mainly diatoms (diatoxanthin) and chlorophytes (lutein) (Leavitt & Hodgson, 2001).

2.5 | Numerical analyses

The Mann–Kendall trend coefficients were used to investigate the first hypothesis that the magnitude of phototrophic assemblage changes was greater in lowland than upland lakes over the last ~200 years. Diagnostic carotenoids from all basin cores ($n = 15$; Supporting information Figure S1a–o) were scaled by mean and variance to account for intersite differences in absolute concentration and preservation from ca. 1800 onwards in dated cores and for the full core length in nondated cores. Principal components analysis (PCA) axis 1 scores were used to summarize changes of the seven main biomarker carotenoids (Supporting information Figure S1a–o). The scaling and PCA were applied using the *vegan* package in R (Oksanen, 2013). All pigments apart from aphanizophyll were strongly correlated with PCA axis 1 in the lowland lakes with the exception of Elterwater's middle basin (low correlations of alloxanthin and zeaxanthin with PCA axis 1) and Windermere's south basin (low correlations of zeaxanthin with PCA axis 1). In the upland lakes, although PCA axis 1 was correlated with alloxanthin in all sites, correlations with other pigments varied among basins (e.g. β -carotene at Blea and Little Langdale Tarns, canthaxanthin at Easedale and Little Langdale Tarns, diatoxanthin at Stickle, Codale and Little Langdale Tarns, lutein in all except Stickle Tarn and zeaxanthin in all except Easedale Tarn) (Supporting information Figure S1a–o).

The Mann–Kendall coefficient, or tau, and p -value for each lake profile ($n = 15$) was determined using the R package *Kendall* (McLeod, 2011). The Mann–Kendall test is a nonparametric test used to determine whether there is a monotonic upward (coefficient between 0 and +1) or downward (coefficient between 0 and –1) trend in a variable over time by calculating a rank correlation coefficient, which is the measure of association between random paired samples in the time series (Figure 2; Supporting information Table S1). This test assigns the sample pairs a score of +1 if the sample is greater than the subsequent samples and –1 if it is lower (Gilbert, 1987). Pigment concentrations sometimes increased in the top few centimetres of each core (see Supporting information Figure S1a–o) due to rapid degradation after recent deposition (Cudington & Leavitt, 1999; Leavitt & Carpenter, 1990). However, because the Mann–Kendall trend test calculates randomly selected pairs of measurements throughout the entire time series, an increase confined to the uppermost centimetres does not significantly influence the dominant trend over time. To determine whether the magnitude of change in primary producer assemblages were significantly greater in lowland (<100 m.a.s.l.) relative to upland lakes (>100 m.a.s.l.), in accordance with hypothesis (1), we conducted dependent t -tests using the R package *vegan* (Oksanen, 2013) on the

Mann–Kendall coefficients based on either PCA axis 1 scores or individual pigments time series.

To address the hypothesis that primary producer communities were regulated mainly by regional factors in upland lakes and local drivers in lowland lakes (hypothesis (2)), regression trees were used to determine the hierarchical relationship among explanatory environmental variables correlated with lake phototrophic assemblage change during the last ~200 years. The analysis was conducted on the dated sediment cores only ($n = 11$). Regression trees are useful tools to detect nonlinear or threshold changes in species and environmental relationships, with the split nodes of explanatory variables chosen according to their ability to minimize the sum of squares error (SSE) (De'ath, 2002). PCA axis 1 scores of the seven biomarker pigments were used as the response variable in the package *mvpart* in R (De'ath, 2002). Each explanatory variable time series differed in length and frequency (described in Supporting information Tables S2 and S3) and so had to be aligned manually to the corresponding pigment time series.

Explanatory variables in the regression tree analysis included changes in individual lake catchment WWTWs and hydrological modifications, livestock and human densities (both individuals/ha), crop type and area coverage (%). Fertilizer application data (available from 1970s onward) were only available at the national level, and climate variables were from a regional dataset (details in Supporting information Tables S2 and S3). Variation inflation factors (VIF) ≤ 10 indicated no collinearity in the explanatory variables (Legendre & Legendre, 2012). The *mvpart()* function was used to perform the regression tree analysis because it avoids overfitting and produced a cross-validation model to predict how well the model performs. The cross-validation model results were plotted and the best-fitted and smallest tree with the lowest associated error (De'ath & Fabricius, 2000) was chosen. To avoid overfitting the model, the tree was pruned according to the one standard error (1-SE) rule, resulting in the smallest tree where the estimated error was within 1-SE of the tree with minimum error (Breiman, Friedman, Olshen, & Stone, 1984). Several cross-validations were run to ensure the 1-SE selected tree remained typical (De'ath & Fabricius, 2000). The percentage variation explained (PVE) by each environmental predictor of primary producer assemblage change was calculated by 1- RE (relative error), and was performed for each single split node. The PVE from each site was then categorized into six environmental variables (further details in supporting information Table S4) and plotted in order of descending lake elevation to understand which variables explained lake primary producer variability across the catchment over the last ~200 years (Figure 3; individual trees provided in Supporting information Figure S2a–k).

Synchrony (S) among lakes was assessed among pairs of fossil pigment records from dated cores ($n = 11$) to test hypothesis (3) that the dynamics in primary producer assemblages were more spatially coherent in upland than lowland basins. To allow comparisons among cores of unequal temporal resolution, scaled pigment concentrations were either averaged where multiple samples existed per 5-year time interval, or linearly interpolated, where missing values arose (Patoine & Leavitt, 2006). This was performed for Grasmere,

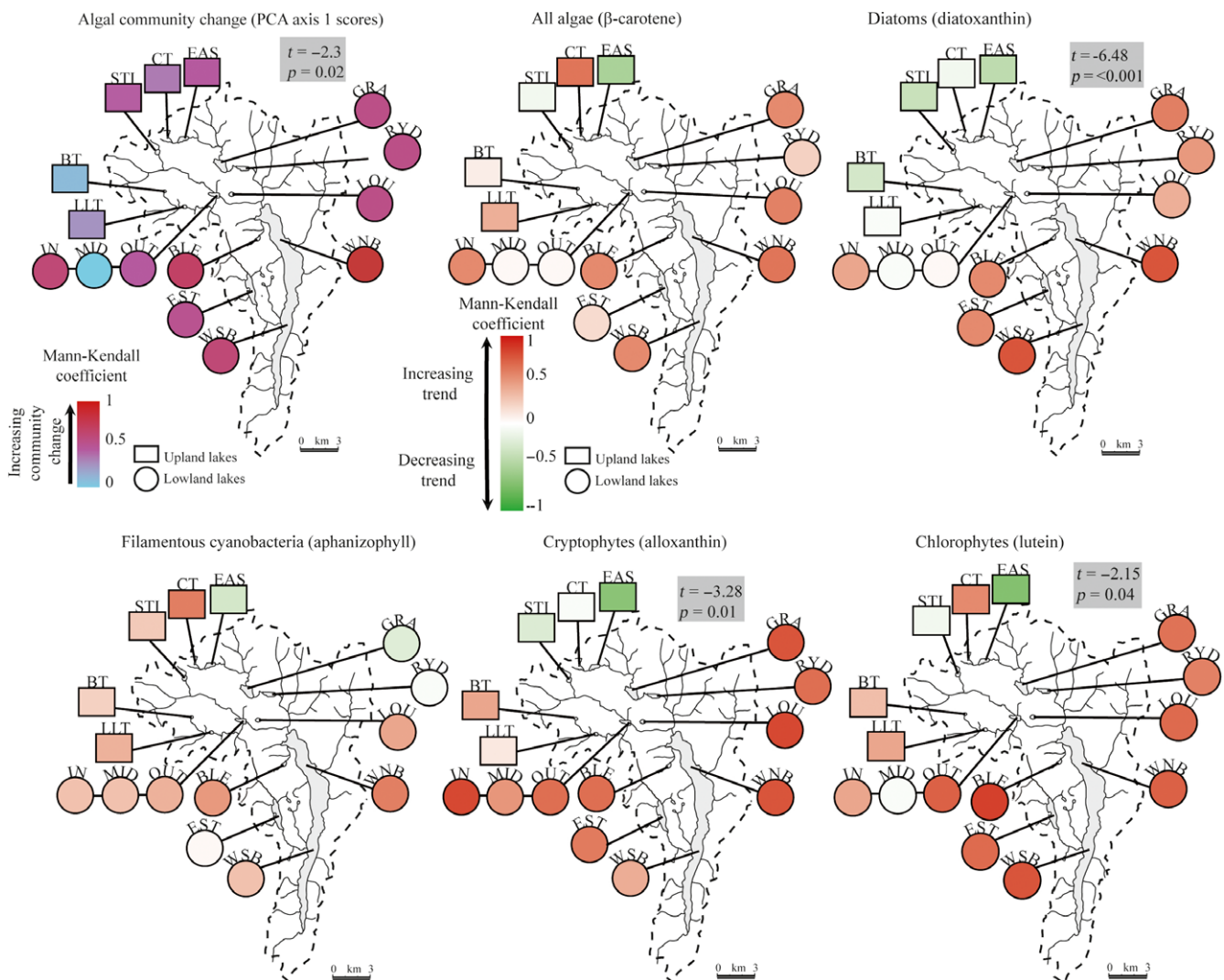


FIGURE 2 Mann-Kendall trends of phototroph assemblage change (PCA axis 1 scores), and individual sedimentary carotenoids from all phototrophs (β -carotene), N_2 -fixing/filamentous cyanobacteria (aphanizophyll), cryptophytes (alloxanthin), diatoms/siliceous algae (diatoxanthin) and chlorophytes (lutein) in lakes of the Windermere catchment over the last ~200 years. Colonial cyanobacteria (canthaxanthin) and all cyanobacteria (zeaxanthin) are given in Supporting information Table S1. The PCA axis 1 scores are presented as absolute values to represent the magnitude of change from no change/zero (light blue) to complete species turnover/one (red). Gradient of red tones indicate positive trends and green indicates negative trends. Abbreviations of lakes: STI, Stickle Tarn; CT, Codale Tarn; EAS, Easedale Tarn; BT, Blea Tarn; LLT, Little Langdale Tarn; LOU, Loughrigg Tarn; EST, Esthwaite Water; GRA, Grasmere; ELTIN, Elterwater inner basin; ELTMID, Elterwater middle basin; ELTOUT, Elterwater outer basin; RYD, Rydal Water; BLE, Blelham Tarn; WNB, Windermere north basin; WSB, Windermere south basin. Significantly ($p < 0.05$) different trend coefficients between upland (squares) and lowland lakes (circles) are reflected in the t-test and p-values presented in the grey boxes

Elterwater inner basin and Windermere's north basin, whose deepest sediment ages were younger than 1800 (1847, 1931 and 1846 respectively), and the upland Codale, Easedale and Stickle Tarns whose resolution was too coarse (11^+ years per 0.5 cm depth at Codale Tarn and 9^+ years from age-depths between 1800 and 1900 at Stickle and Easedale Tarns). The synchrony of six scaled taxon-specific carotenoids (as above) and total algal abundance (as β -carotene) during the period 1800–2005 was determined in the R package *synchrony*. For individual pigments, the synchrony value is the mean Pearson correlation coefficient (r) of a group of lake pairs (Gouhier & Guichard, 2014; Kratz et al., 1998; Vogt, Rusak, Patoine,

& Leavitt, 2011). Monte Carlo permutation testing was used to establish the statistical significance ($p \leq 0.05$) of each synchrony value. All time series were truncated at 2005. High levels of regional synchrony ($S > 0.5$) suggest that regional drivers such as climate or catchment-wide land use practices exert a dominant control on the response variables (George et al., 2000; Patoine & Leavitt, 2006). The results of the regression tree analyses described above implied timing of WwTWs varied among the lowland lakes ($n = 7$), so synchrony analysis was performed on these lakes following the onset of point nutrient activity (1886) to help interpret the synchrony patterns in these impacted basins.

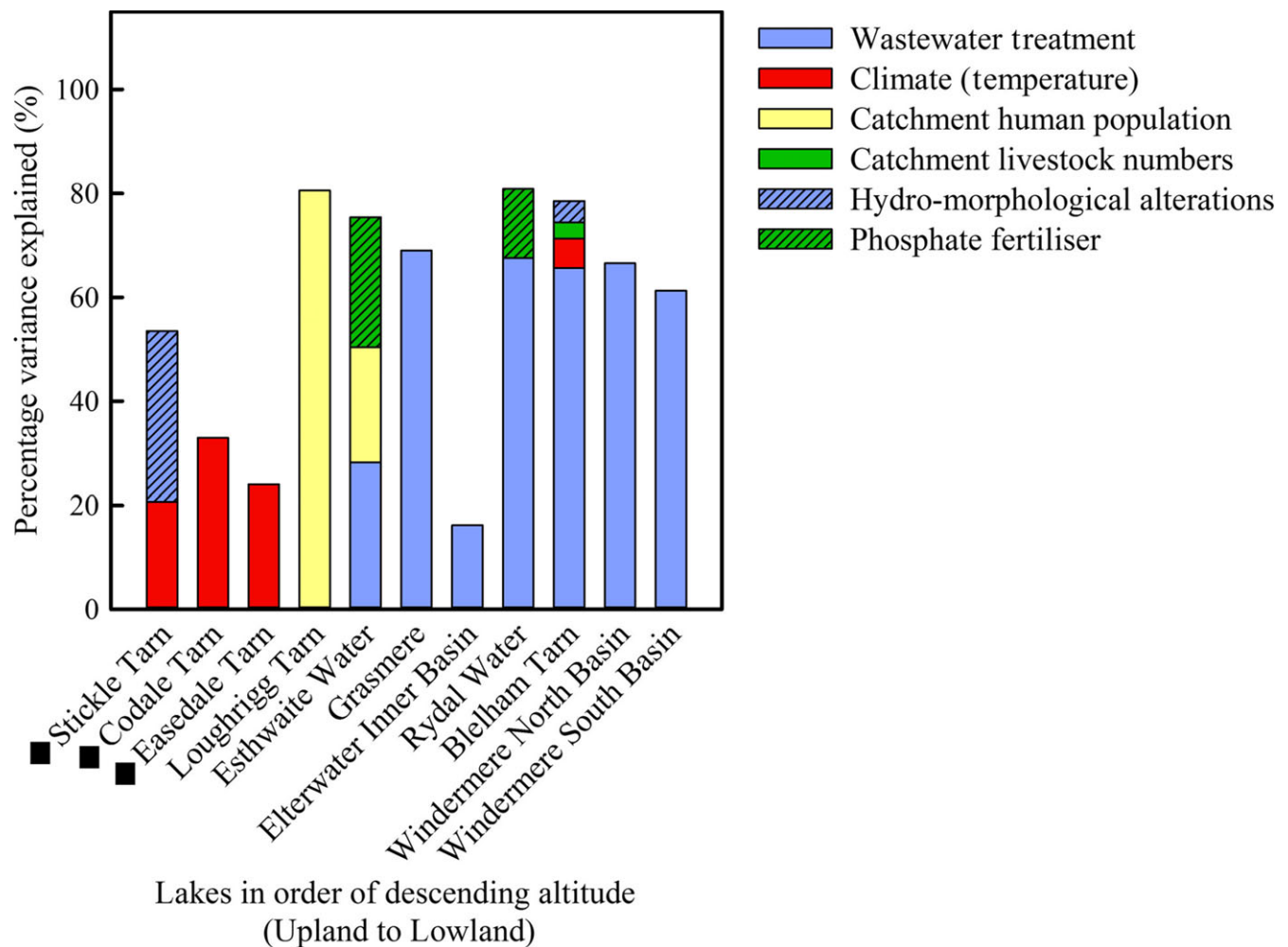


FIGURE 3 Percentage variance explained by the environmental variables selected by regression tree analysis as significant predictors of algal assemblage change (PCA axis 1 scores) in order of descending lake altitude. Squares are shown next to upland lakes (>100 m.a.s.l.). Further details on environmental variables given in supporting information Tables S2 and S3, and on the regression trees in Supporting information Figure S2a-k, Supporting information Table S4

Finally, to examine the concept of lake and their catchments characteristics as “filters” of lake response (Blenckner, 2005), the Pearson's correlation coefficients (r) from the seven carotenoids for catchment-wide lake pairs (1800–2005) were used as the response variable in a redundancy analysis (RDA) with noncollinear ($VIF < 10$) lake and catchment characteristics (difference in altitude, maximum depth, water retention time (WRT), catchment to lake area ratio (CA: LA), geology between lake pairs) as the explanatory variables (Figure 4). Continuous explanatory variables were first scaled by mean and variance to control for differences in magnitude prior to running the RDA. Both the scaling and RDA were undertaken in R using the *vegan* package (Oksanen, 2013).

3 | RESULTS

Concentrations of sedimentary pigments increased over the last ~200 years, as determined by positive Mann–Kendall trend values for most of the lake time series (Figure 2; Supporting information

Table S1). The magnitude of trends in PCA axis 1 scores was greater for all lowland lakes than for upland lakes, except for the lowland Elterwater middle basin. Lowland lakes (Grasmere, Rydal Water, Blelham and Loughrigg Tarn, Esthwaite Water and Elterwater and Windermere basins) had predominantly positive, increasing trends in individual pigments. In contrast, trends varied between positive and negative in the upland lakes, with notable negative, decreasing trends for all algal pigments at Easedale Tarn. Distinct differences between upland and lowland lake trends in mainly diatom (diatoxanthin), cryptophyte (alloxanthin) and chlorophyte (lutein) pigments were found supporting hypothesis (1) that assemblage change was greater in the lowlands.

Regression tree analyses indicate that timing of the establishment and upgrades of WwTWs explained the greatest amount of variation in primary producer assemblages over the last ~200 years across the entire Windermere catchment (Figures 3, Supporting information Figure S2a-k; Supporting information Table S4). Overall, changes in WwTWs predicted the assemblage change at the most lakes ($n = 7$); however, it is notable that this result was restricted to

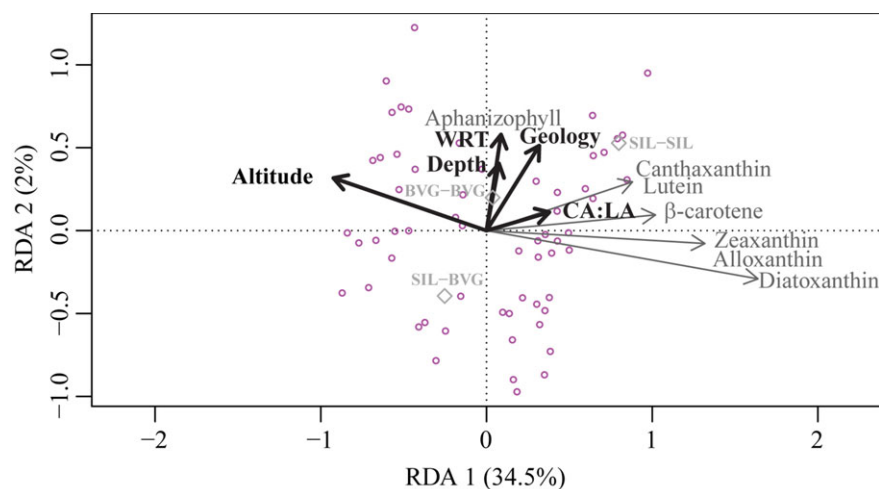


FIGURE 4 Redundancy analysis exploring relationship between the Pearson's correlation coefficients (r) of lake pairs' seven sedimentary pigments from dated sediment cores (1800–2005, $n = 11$) as the response variables, and the difference in lake and catchment characteristics for these Windermere catchment lake pairs as the explanatory variables. The r values were both positive ($0-1^+$) and negative (1^-0) (where negative correlations occur when pigment concentrations at one site go up and at the other go down). The position of the pigment labels reflect the extent to which the changes in synchrony contribute to the canonical relationships. Centroids reflecting the geological classes in each of the lake pairs are shown in grey (SIL, Silurian slate geology; BVG, Borrowdale volcanic group geology). Other abbreviations used are: WRT, water retention time and CA:LA, catchment to lake area ratio

lowland lakes, whereas climate was a greater predictor of change in uplands as hypothesized (hypothesis 2) (Figure 3). Interestingly, human population density was the most important predictor of phototrophic communities at the lowland Loughrigg Tarn (PVE = 81%), a basin which is comparatively isolated from WwTWs and other surface inflows. Temperature was the most important correlate of changes in primary producer assemblages at two upland lakes Codale (PVE = 33%) and Easedale Tarns (PVE = 24%), and hydro-morphological alteration was the main predictor at upland Stickle Tarn (PVE = 33%), followed by temperature (PVE = 21%).

Historical variation in abundance of primary producers was more synchronous between lowland (S range 0.16–0.69; $p < 0.001$) than between upland basins where pigment synchrony values were non-significant (Table 2). Synchrony values of most pigments were relatively high ($S \geq 0.49$) between lowland lakes, with the exception of aphanizophyll which had a lower but still significant ($p > 0.001$) value of 0.16. Sedimentary pigments were more synchronous (S range 0.3–0.61; $p < 0.001$) prior to the onset of WwTW activity across the lowland lakes compared to afterwards which supports hypothesis (3); that basin-specific activity would increase asynchrony in the

lowland lakes. However, unlike the other carotenoids, cryptophyte and filamentous cyanobacteria pigments (alloxanthin and aphanizophyll respectively) had lower synchrony before the onset of WwTW development and higher synchrony afterwards.

The RDA of the Pearson's correlation coefficients (r) from the seven carotenoids constrained to the environmental variables showed that pigment synchrony varied due to lake and catchment characteristics. In particular, the first RDA axis, which was significantly and positively correlated with the original r correlation coefficients, showed that the synchrony of primarily diatom (diatoxanthin), followed by cryptophyte (alloxanthin) and total cyanobacterial (zeaxanthin) pigments was most strongly correlated with differences in altitude (axis 1 = 34.5% variance explained), with greater synchrony when differences in elevation between lake pairs were smaller (Figure 4). Synchrony patterns of chlorophyte (lutein) and colonial cyanobacteria (canthaxanthin), followed by total algae (β -carotene) were closely correlated with changes in catchment to lake area ratios, although this was a lower contributor to the variance explained. Synchrony of the filamentous cyanobacteria (aphanizophyll) was correlated with differences in lake-specific factors such as

TABLE 2 Mean synchrony values for seven carotenoids across all lake pairs of the Windermere catchment in upland and lowland lakes and across all lowland lake pairs of the Windermere catchment before and after Wastewater Treatment Work establishment and development

	Alloxanthin (cryptophytes)	Aphanizophyll (filamentous cyanobacteria)	β -carotene (all algae)	Canthaxanthin (colonial cyanobacteria)	Diatoxanthin (mainly diatoms)	Lutein (chlorophytes)	Zeaxanthin (all cyanobacteria)
Upland lakes (>100 m.a.s.l.)	−0.13	−0.06	−0.01	−0.2*	0.12	−0.27	−0.07
Lowland lakes (<100 m.a.s.l.)	0.68**	0.16**	0.49**	0.58**	0.60**	0.69**	0.61**
Before WwTW (1800–1885)	0.13*	0.08	0.31**	0.3**	0.3**	0.54**	0.61**
After WwTW (1985–2005)	0.37*	0.46*	0.22*	−0.04	−0.06	−0.14	−0.04

* $p \leq 0.05$, ** $p \leq 0.001$.

maximum depth, water retention time, and to a lesser extent geological class, although variance explained along this axis was small (axis 2 = 2% variance explained).

4 | DISCUSSION

Across the majority of lakes in the Windermere catchment, we observed increasing concentrations of ubiquitous pigments (β -carotene) over the last ~200 years, indicating a catchment-wide increase in lacustrine primary production since 1800 (Figure 2). However, in agreement with hypothesis (1), the trends towards higher abundance were most distinct in lowland lakes (<100 m.a.s.l.) where human activities have been most intense, as compared with basins at higher elevations. Regression tree analysis supported hypothesis (2) and showed that local drivers (WwTW installations) explained most variance in lowland lakes, whereas regional factors (temperature) were the dominant drivers at uplands sites (Figure 3). In contrast with hypothesis (3), however, temporal coherence among pigments in lowland lakes was greater than that observed among upland sites over the last ~200 years (Table 2).

4.1 | Lowland lakes

This study demonstrated that lowland lakes exhibited the greatest increase in the abundance of algae and cyanobacteria within the Windermere catchment since 1800 reflecting effects of intense individual subcatchment anthropogenic disturbance (hypothesis (1), Figures 2 and 3). Significant trends in PCA axis 1 scores also indicate that these increases in primary producer abundance were accompanied by changes in assemblages that themselves were highly synchronous across lowland sites (hypothesis (3), Table 2). Consistent with earlier work on individual lakes (Dong et al., 2012; McGowan et al., 2012; Moorhouse et al., 2014), the major driver of assemblage change (as PCA axis 1 scores) appeared to be the installation and upgrade of WwTWs (hypothesis (2), Figure 3). As shown in other northern temperate lakes, influx of nutrients, particularly P from point sources, commonly accelerates eutrophication of oligotrophic and mesotrophic lakes (e.g. Edmondson, 1970; Jenny et al., 2016; Schindler, 2006).

The observation that the timing of WwTW operation is more important for altering the production and composition of phototrophs than the growth of resident human populations, may reflect the fact that major tourist towns are located within subcatchments of lowland lakes (Figure 1) and that the >15 million annual visitors are excluded from population estimates yet drive a need for WwTW development. We note that in the absence of WwTWs, human population densities within the rural Loughrigg Tarn subbasin explained almost 80% of the variability in algal assemblage change (Figures 3 and S2d; Supporting information Table S2–S4). Here, nutrient inputs from humans are derived mainly from septic tank systems. Although septic systems were also common elsewhere in the lowlands of the catchment, particularly in the 19th century (McGowan et al., 2012),

the funnelling of waste into WwTWs with insufficient nutrient removal can mask the diffuse nutrient effects of septic tank systems (Reynolds, Maberly, Parker, & De Ville, 2012), potentially explaining why it was a weaker predictor of phototrophic change in lakes with point WwTWs. However, such diffuse nutrient systems may explain the synchronous changes in primary producers in lowland lakes during the 19th century, prior to asynchronous WwTW activity (Table 2). Finally, we note that the hydro-morphological change at Blelham Tarn (i.e. channelization of inflow) (Figures 3 and S2i; Supporting information Table S4) has previously been associated with increasing efficiency of transport of nutrients from the WwTW into the lake (Moorhouse et al., 2014). Taken together, our findings highlight the overwhelming imprint of human wastes on all lowland lakes within the Windermere catchment.

We found limited support for the expectation that regional intensification of agriculture since 1800 is an important control of abundance and gross compositional modifications of primary producers (Keatley et al., 2011). For example, synchrony of lowland lake pairs prior to the onset of WwTWs (early-mid 19th century) tended to be higher than that recorded after point urban nutrient influxes increased (Table 2), possibly reflecting regional growth in ploughing, deforestation and mechanization of agriculture in the lowlands during the 19th century (McGowan et al., 2012; Pennington, Cambray, Eakins, & Harkness, 1976). However, local land use practices were generally weak predictors of phototrophic assemblage change. For example, livestock explained 3.1% of the variance in Blelham Tarn pigment composition, despite marked increases in sheep rearing since the mid-1800s, and consequent promotion of soil erosion in this subbasin (Van Der Post, Oldfield, Haworth, Crooks, & Appleby, 1997). Similarly, while changes in P fertilizer explained 13.3% and 25% of variance in pigment composition from Rydal and Esthwaite Waters respectively, their variance explained was still less than WwTWs and contradicts the importance of fertilizer nutrients in other agricultural catchments (Bunting, Leavitt, Gibson, Mcgee, & Hall, 2007; Donald, Bogard, Finlay, Bunting, & Leavitt, 2013). Thus, although basin-specific estimates of fertilizer application were not available, and could have been collinear with other parameters (livestock or human density) (Bunting et al., 2016), our findings generally imply that influx of human wastes, rather than agricultural production, was the main factor degrading lotic lowland ecosystems, particularly since the onset of the 20th century.

4.2 | Upland lakes

In contrast to the lowland sites, Mann–Kendall trends in upland lakes were weak and nonsignificant for several basins (Figure 2; Supporting information Table S1). Lack of consistent trends most likely reflects the absence of human settlements, minimal farming at elevation (Pickering, 2001; Tipping et al., 1998), and reduced importance of nutrient influx on water quality. The absence of strong synchrony among upland basins is unexpected, however, given the paramount effect of temperature (20%–30% of variance along PCA axis 1) in three upland lakes (Stickle, Codale, Easedale Tarns) (Figure 3), a

nearly 1°C increase in regional mean temperatures since 1987 (Dong et al., 2012), and the observation that temperature variability often increases temporal coherence among lakes within even large lake watersheds (Leavitt et al., 2009; Magnuson, Benson, & Kratz, 1990; Vogt et al., 2011).

4.3 | Upland and lowland lake synchrony

Greater overall synchrony of the lowland lakes relative to the upland lakes may arise in part because of the greater connectivity of the lowland lakes (e.g. lower order in lake chain) via surface and groundwater flows and a greater degree of cumulative influence of nutrient inputs (Kratz et al., 1998). Lakes within a chain often have higher synchrony of limnological variables relative to headwater or spatially isolated basins that are influenced more by precipitation than surface inflows (Kratz, Webster, Bowser, Magnuson, & Benson, 1997; Riera, Magnuson, Kratz, & Webster, 2000; Soranno et al., 1999). In addition, isolated lakes tend to be more hydrologically complex, leading to more individualistic responses to climate forcing (Webster et al., 2000). Overall, our study contributes to the general theory that lake position within the hydrological landscape is a critical control of its response to forcing by climatic and anthropogenic processes (Soranno et al., 2009).

Site-specific differences in precipitation may have reduced the coherence of primary producers in upland lakes despite potentially synchronizing effects of regional climatic change (Figures 3 and 4). For example, given that regional precipitation and snow/ice cover patterns are strongly influenced by orographic processes and therefore, lake position in the catchment (George et al., 2007), and that the influx of precipitation can reduce limnological coherence within lake catchments (Vogt et al., 2011), we infer that spatial variability in rainfall (Malby, Whyatt, Timmis, Wilby, & Orr, 2007) may have led to regionally asynchronous delivery and removal of nutrients in the relatively pristine upland sites, further modified by basin-specific lake flushing (Maberly et al., 2002; Soranno et al., 2014).

Our results show a hierarchal scale of landscape (altitude, catchment to lake area ratios) followed by basin (water retention times, maximum depth) characteristics on driving synchrony dynamics of algal assemblages over the last ~200 years (Figure 4), supporting the notion that such characteristics “filter” a lakes response to environmental change (Blenckner, 2005). This corresponds to previous work, which found that landscape factors (i.e. altitude) often define and constrain the basin-scale factors (Soranno et al., 2009). Interestingly, the RDA showed that once the effects of landscape were partialled out, lake depth and water residence time accounted for additional variability in pigment synchrony. In particular, lake depth and water residence time were closely associated with the synchronous dynamics of filamentous cyanobacteria (aphanizophyll), which have been shown to be important factors in shaping cyanobacterial communities elsewhere (Posch, Koster, Salcher, & Pernthaler, 2012). Considering the detrimental impacts of cyanobacterial blooms on human and ecological health (Paerl & Huisman, 2009), and predictions that this group will increase with increasing temperatures and decreased flushing rates (Elliott, 2010), our findings highlight the importance of

extricating cyanobacterial dynamics in accordance with the “filtering” effect of basin characteristics. Thus, while we conclude that the proximity of a lake to anthropogenic activity is a key driver of lake primary production modifications; basin-specific characteristics alter this relationship further.

4.4 | Implications for water management

Changes in primary producer assemblages were greatest in lowland lakes receiving influx of human wastewater. In particular, the terminal basins of Lake Windermere integrated pollutants throughout the catchment and displayed the highest magnitude of algal modifications (Figure 2). Strong effects of nutrients on lake production are noted regularly elsewhere (Bunting et al., 2016; Leavitt et al., 2009; Scheffer, Hosper, Meijer, Moss, & Jeppesen, 1993; Taranu et al., 2015) and have been suggested to override the effects of climate change in other lake catchments (Leavitt et al., 2009; Moorhouse et al., 2014; Taranu et al., 2015; Vogt et al., 2011). Based on these findings, it is clear that management of Lake Windermere requires a consideration of other upstream basins, in particular strategies to reduce emissions from WwTWs. Furthermore, the lack of a common trend in lake production exhibited by headwaters highlights the need to improve our understanding of these ecosystems and not overstate their resilience to continued global warming.

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SUPPORTING INFORMATION

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